TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. 371

ATTORNEY'S DOCKET NUMBER

19380.0009

U.S. APPLICATION (If known, see 37 CFR 1.5) 8 9 7 4 3

INTERNATIONAL FILING DATE PRIORITY DATE CLAIMED INTERNATIONAL APPLICATION NO. 12 January 2000 27 January 1999 PCT/FI00/00018 TITLE OF INVENTION PHYSIOLOGICAL FOOD SALT PRODUCT APPLICANT(S) FOR DO/EO/US Juhani Mäki Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information: 1. [x] This is a FIRST submission of items concerning a filing under 35 U.S.C. 371 This is a SECOND or SUBSEQUENT submission of items concerning a filing under 35 U.S.C. § 371. This express request to begin national examination procedures (35 U.S.C. 371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39(1). A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date. 5. [x] A copy of the International Application as published (35 U.S.C. 371(c)(2)) WO 00/44245 [x] is transmitted herewith (required only if not transmitted by the International Bureau). has been transmitted by the International Bureau. is not required, as the application was filed in the United States Receiving Office (RO/US). 6. A translation of the International Application into English (35 U.S.C. 371(c)(2)). Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3)) are transmitted herewith (required only if not transmitted by the International Bureau). ☐ have been transmitted by the International Bureau. have not been made; however, the time limit for making such amendments has NOT expired. have not been made and will not be made. 8. \(\sigma\) A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3). 9. [x] An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)). 10. 🗆 A translation of the Annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)). Items 11. to 16. Below concern other document(s) or information included: [x] An Information Disclosure Statement under 37 CFR 1.97 and 1.98, PTO-1449, 8 references 11. 12. ☐ An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included. [x] A FIRST preliminary amendment. 13. A SECOND or SUBSEQUENT preliminary amendment. 14. A substitute specification. 15. A change of power of attorney and/or address letter 16. [x] Other items or information: PCT/ISA/210 PCT/IB/332 PCT/IPEA/409

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U.S. PPLICATION NO. (If kn	own, see 37 CFR 15) 89743	INTERNATIONAL APPLICATION NO. PCT/FI00/00018		ATTORNEY'S DOCKET NUMBER 19380.0009	
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Surcharge of \$130.00 for the earliest claimed priori			20 [x] 30 months from	\$	
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Total Claims	18 - 20 =	0	X \$18.00	\$	
Independent Claims	4 - 3 =	1	X \$80.00	\$ 80.00	
Multiple dependent clain	n(s)(if applicable)		+ \$270.00	\$	
TOTAL OF ABOVE CALCULATIONS =				\$1,080.00	
Reduction by 1/2 for filing by small entity, if applicable.			\$ 540.00		
			SUBTOTAL =	\$ 540.00	
Processing fee of \$130.00 for furnishing the English translation later than \square 20 \square 30 months from the earliest claimed priority date (37 CFR 1.492(e)).			\$		
TOTAL NATIONAL FEE =				\$ 540.00	
Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). \$40.00 per property +			\$		
TOTAL FEES ENCLOSED =			\$ 540.00		
				Amount to be: Refunded	
				Charged	\$ 540.00
a. A check in the an	nount of \$ to cover the	e above fees is enclosed.			

- b. [x] Please charge my Deposit Account No. 19-5127, Order No. 19380.0009 in the amount of \$ 540.00 to cover the above fees. A duplicate copy of this sheet is enclosed.
- c. [x] The Director is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. 19-5127. A duplicate copy of this sheet is enclosed.

NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or (b) must

be filed and granted to restore the application to pending status SEND ALL CORRESPONDENCE TO:

Edward A. Pennington

Swidler Berlin Shereff Friedman, LLP

3000 K Street, N.W., Suite 300

Washington, DC 20007-5116

SIGNATURE

Eric J. Franklin

NAME

37,134

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Application of: :

Juhani Mäki

•

Serial No. -- Corresp. to PCT/FI00/00018 : Art Unit: -

Filed: July 20, 2001 : Examiner: -

For: PHYSIOLOGICAL FOOD SALT PRODUCT

PRELIMINARY AMENDMENT

Assistant Commissioner for Patents Washington, DC 20231

Sir:

Prior to examination, please amend the above-identified application as follows:

In the Claims:

Clean copy of the amended claims:

- 3. The product according to claim 1, **characterized** in that the magnesium ammonium chloride and/or the calcium ammonium chloride is in a complex form.
- 5. The product according to claim 1, **characterized** in that the general anhydrous formula of the salt contained in the product is $aMg \times bCa \times NH_4Cl_3$, in which a+b=1, and a and b are greater than 0, and in which part of the ammonium can be replaced with potassium.
- 6. The product according to claim 1, **characterized** in that the general anhydrous formula of the salt contained in the product is in the type $MgNH_4Cl_3 \times eCaCl_2$, in which e is preferably not greater than 0.2 and in which part of the ammonium can be replaced with potassium.

- 7. The product according to claim 1, **characterized** in that the general anhydrous formula of the salt contained in the product is $Mg \times cNH_4 \times dK \times Cl_3$, in which c+d=1, and c and d are greater than 0, preferably so that $c \ge 0.5$.
- 8. The product according to claim 1, **characterized** in that it contains sodium chloride and/or potassium chloride.
- 10. The product according to claim 1, **characterized** in that it contains materials which are advantageous to vital functions, such as micronutrients, vitamins, flavonoids, steroids, or the like.
- 11. The product according to claim 1, **characterized** in that it contains as additives affecting primarily the taste of the product carbohydrates or their polymeric forms, spices, herbs, acidity regulators, glutamates, proteins, protein hydrolysates, or the like.
- 17. The method according to claim 13, **characterized** in that the pH of the mother liquor is adjusted by means of a hydroxide, particularly potassium or ammonium hydroxide, particularly to adjust the crystallization of free ammonium chloride.

Amended Claims

- 3. (Amended) The product according to claim 1 [or 2], **characterized** in that the magnesium ammonium chloride and/or the calcium ammonium chloride is in a complex form.
- 5. (Amended) The product according to claim 1 [or 2], **characterized** in that the general anhydrous formula of the salt contained in the product is $aMg \times bCa \times NH_4Cl_3$, in which a + b = 1, and a and b are greater than 0, and in which part of the ammonium can be replaced with potassium.
- 6. (Amended) The product according to claim 1 [or 2], **characterized** in that the general anhydrous formula of the salt contained in the product is in the type MgNH₄Cl₃× eCaCl₂, in

which e is preferably not greater than 0.2 and in which part of the ammonium can be replaced with potassium.

- 7. (Amended) The product according to claim 1 [or 2], **characterized** in that the general anhydrous formula of the salt contained in the product is $Mg \times cNH_4 \times dK \times Cl_3$, in which c+d=1, and c and d are greater than 0, preferably so that $c \ge 0.5$.
- 8. (Amended) The product according to [any of the preceding claims] <u>claim 1</u>, **characterized** in that it contains sodium chloride and/or potassium chloride.
- 10. (Amended) The product according to [any of the preceding claims] claim 1, **characterized** in that it contains materials which are advantageous to vital functions, such as micronutrients, vitamins, flavonoids, steroids, or the like.
- 11. (Amended) The product according to [any of the preceding claims] <u>claim 1</u>, **characterized** in that it contains as additives affecting primarily the taste of the product carbohydrates or their polymeric forms, spices, herbs, acidity regulators, glutamates, proteins, protein hydrolysates, or the like.
- 17. (Amended) The method according to [any of the preceding claims 13 to 16] <u>claim 13</u>, **characterized** in that the pH of the mother liquor is adjusted by means of a hydroxide, particularly potassium or ammonium hydroxide, particularly to adjust the crystallization of free ammonium chloride.

Date: July 20, 2001

Remarks

Applicants have amended the claims to eliminate multiple dependencies to reduce the filing fee.

Respectfully submitted,

Eric J. Franklin, Reg. No. 37,134

Swidler Berlin Shereff Friedman, LLP

3000 K Street, N.W., Suite 300 Washington, DC 20007-5116

Physiological food salt product

The present invention relates to a physiological food salt product which primarily consists of minerals and in whose formulation an alkaline earth metal component or components are brought to an appropriate form with acceptable taste and hygroscopic properties. The invention also relates to the use of a salt product prepared according to the

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method.

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In the preparation, preservation or seasoning of foods in a variety of cases, with respect to the use of food salt products, it is presently an important aim to avoid excessive inclusion of common salt, *i.e.* sodium chloride, NaCl, in the formulation of the food product. The reason for this attempt is the clearly documented harmful effect of NaCl on human health. When the daily consumption of NaCl by a person in the Western countries is estimated to be 170 meq (9.9 g), morbidity on arterial hypertension and mortality on myocardial infarction are common even with daily intake of 100 meq (5.8 g). When the daily dose is less than 50 meq (2.9 g), morbidity is indicated to be low. Consequently, the above-mentioned value 50 meq per day can be considered the recommended maximum daily intake of NaCl.

It has been found in different cases that a daily intake of magnesium,

Mg, reduces blood pressure. The recommended dietary allowance
(RDA) indicated for magnesium is 4.5 mg/kg/day, resulting in calculated
values of 350 to 400 mg per day for adult male population and 280 to
300 mg per day for women, correspondingly. For the above-mentioned
reasons, there is a strong need to develop a substitute or modification
for common salt, which will fulfill not only the physiological recommended values but also other requirements, such as taste and good
sprinklability in its most common form of usage.

The RDA value for calcium, Ca, is indicated to be 800 to 1200 mg per day for adult population. Although the supply of calcium is often sufficient, particularly when the diet is based on milk products, it can be

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considered appropriate to include calcium in table salt, for example due to an increased need of Ca during pregnancy or during adolescence.

To combine physiologically sufficient alkaline earth contents with food salt formulations has been problematic in practice, primarily because other salts than chlorides (sulphates, phosphates, carbonates, etc.) can hardly be used because of their physiological unsuitability, bitter taste and/or poor solubility.

When alkaline earth metal chlorides are added to food salt in connection with the preparation process, one must, however, solve in a satisfactory manner their strong hygroscopicity, which means the tendency of deliquescence of the product under the effect of humidity in the air. Furthermore, the taste of these chlorides is pungently salty and thus too different from the taste of NaCl.

The same properties apply to carnallite, $MgCl_2 \times KCl \times 6H_2O$, present in natural salt deposits which normally contains such quantities of coprecipitated bromides that its nutritional use as such is out of question.

Upon studying the prior art related to the invention, it is found that the development of table salt products has recently been focused on products containing potassium, K, or potassium and magnesium, K and Mg, whereby the main purpose is to reduce the intake of sodium, Na, in food, the main criteria for commercial utilization being acceptable taste. Consequently, methods have been introduced to prepare such mixtures of NaCl, KCl and MgCl2 in which MgCl2 or its potassium double salt imitating the natural carnallite is protected from air humidity by means of alkali metal chlorides. Two patent applications that are close to these subjects should be mentioned, WO A1 92/16117 (A23L 1/237) and WO A1 92/18668 (A23L 1/237). Of these, the former presents a method for coating the hygroscopic component of the salt with a non-hygroscopic material. The latter patent application presents a method for treatment of NaCl-K/Mg salts, which method comprises quick drying of the solution prepared of the salts and is intended for producing a stable multicomponent salt mixture.

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Patent application WO 90/00522 (A23L 1/237) describes a synthetic method for preparing a salt which imitates carnallite for a food salt product by evaporating a solution which contains potassium chloride and magnesium chloride in equivalent ratios and by heat treatment of the obtained evaporation residue to stabilize the product. However, it is industrially impractical to remove scale deposits from the evaporating apparatus, and the method cannot thus be realized as a continuous process. Moreover, it has been found in check-up studies relating to the method that a dried evaporation residue obtained with e.g. a rotatory evaporator is as hygroscopic as natural carnallite, and that as a result of the heat treatment, hydrochloric acid is emitted from the product, which results in that the solubility of the product to water, the pH of the solution, and thereby its taste are changed to the adverse direction.

Aalso patent application WO 88/09131 (A23L 1/237) should be mentioned as related to the invention. The application presents a salt substitute which contains 50 % of free ammonium chloride NH₄Cl admixed mechanically to phosphates and sugar which adjust the acidity of the product. Continuous use of the product according to the formulation may, however, be physiologically doubtful, because the calculatory ammonium content will thus rise to a level of 16.8 %.

Furthermore, as prior art connected with the invention, two Finnish patent applications should be mentioned, No. 961229 and No. 970323, as well as international application WO 98/32343 (A23L 1/237) related to the latter, in which, to eliminate the hygroscopicity, an amino acid complex is formed from alkaline earth chlorides, particularly with glycine, which is also the simplest amino acid. However, a closer study of the technique presented in the patents has revealed that the aims of the patents cannot be achieved to the described extent, for the following reasons.

For example, $MgCl_2 \times 6H_2O$ forms three different complexes with glycine, which have been found to be in a balance with each other according to the following equation:

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K' K'' K''' K''' $M \times H_6 \leftrightarrow M \times L \times H_4 \leftrightarrow M \times L2 \times H_2 \leftrightarrow M \times L$

 $H = H_2O$ L = Glycine $M = MgCl_2$ k = equilibrium constant

For example, a compound prepared with equivalent ratios of MgCl₂ and glycine is thus normally a mixture which does not fulfill the criterion of hygroscopicity at high air humidity values. Moreover, it is not possible to isolate amino acid complexes from a concentrated solution by centrifugation, but the solution must be fully evaporated, which complicates

industrial production. Furthermore, it seems that adverse changes take place in the taste and colour of the final product even in a short time interval, possibly due to oxidation of the organic component.

In any case, none of the products according to any of the above-mentioned applications fulfills all the requirements set above for the invention. The same applies to commercially available Na-K-Mg table salts, examples of which include the almost identical products Seltin® and Pan Salt® whose composition comprises the compounds NaCl, KCl and MgSO₄ \times 7H₂O (12 %) in a mechanical mixture. When the intake of magnesium is calculated from e.g. the above-mentioned commercial products with an average daily use of 7.5 g, the daily dose obtained for magnesium is 88 mg, which is fully insufficient in view of the above-mentioned recommended values. In these products, the hygroscopic

properties are under control, but it has not been possible to eliminate the disadvantages of the sulphate, of which an increased excretion of calcium possibly caused by the sulphate ion should be mentioned. Furthermore, analyses of the compositions of both products have revealed MgSO₄ contents differing from the given values, which may be primarily due to the fact that this salt has a different crystalline form and density than the alkali metal chlorides, which results in the separation of

It is disclosed in prior art that ammonium carnallite type compounds of alkaline earth metals, particularly the corresponding magnesium salt, have been used for preparing anhydrous alkaline earth chlorides by a pyrolytic method for technical purposes, of which the most important is

magnesium sulphate from the mixture.

the electrolytic preparation of magnesium metal from MgCl₂ melt in a process which is now outdated (British patent 351 845 from the year 1931 and German Offenlegungsschrift 1 567 937 from the year 1970). The uses in question do not, however, fall into the category of the present application, and the patent publications in question do not discuss the hygroscopic properties of the prepared products.

The purpose of the invention is to introduce a method, by which it is possible to eliminate the above-mentioned drawbacks related to the preparation of food salts and to make a product containing the desired ions, particularly alkaline earth metal chlorides, which fulfills the requirement of sufficiently low hygroscopicity as well as makes it possible to achieve the objectives related to the taste and nutritional properties set for the salt product.

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To achieve the above-mentioned aims, the invention is primarily characterized in what will be presented in the characterizing part of the appended claim 1. By means of ammonium chloride salts of the alkaline earth metals Mg and Ca, it is possible to produce a nutritional salt product which contains the desired ions but whose physical properties are also suitable for practical applications.

The method of the invention for preparing physiological food salts is based on the surprising finding that when ammonium chloride is added into substantially concentrated solutions, normally aqueous solutions, which contain alkaline earth metal chloride or chlorides, and can preferably also contain potassium chloride and possible other components and/or micronutrients affecting primarily the taste, it is possible to obtain, particularly in the presence of magnesium chloride, carnallite-type crystalline precipitations which fulfill the above-mentioned requirements relating to low hygroscopicity and the taste. Furthermore, the taste, primarily the acidity, of the obtained carnallite-type salts can be influenced preferably by changing the pH value of the mother liquor with bases, preferably ammonia, and/or by treating the product separated from the mother liquor at suitably selected temperatures.

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With reference to the invention, it has been surprisingly found that ammonium carnallite, whose formula can be given either as $MgCl_2 \times NH_4Cl \times 6H_2O$ or $MgNH_4Cl_3 \times 6H_2O$, has the particular property to coprecipitate hygroscopic chlorides, such as calcium chloride $CaCl_2$ or potassium carnallite $MgKCl_3 \times 6H_2O$ from the mother liquor to mixed crystal forms which have no hygroscopicity or in which the property is substantially suppressed.

The use and modification of ammonium chloride hydrates of alkaline earth metals in the above-described manner to prepare the present physiological food salt product and to control its properties is not disclosed in the prior art. It is thus a new inventive idea.

The inclusion of hygroscopic alkaline earth metal chlorides or their double salts, such as potassium carnallite, in sprinklable food salt products has been previously difficult or almost impossible. These impediments can be eliminated by forming a crystalline ammonium chloride adduct from the salts.

In the following, the invention and the related technique will be described in more detail with reference to the appended graphs, in which

Fig. 1 shows the dependence of NH₄ content in ammonium carnallite, used as an exemplary product, on the quantity of water used for preparing the mother liquor,

Fig. 2 shows the dependence of pH value of ammonium carnallite, used as an exemplary product, on the quantity of 5 % ammonium hydroxide added per mole in the mother liquor, and

Fig. 3 shows the effect of heat treatment on the pH value of two ammonium carnallite products prepared by different methods, as a function of time.

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The following is a more detailed description on the properties of food salt components containing alkaline earth metals, relating to the invention, as well as on the preparing technique related to the invention.

In studies related to the invention, it has been surprisingly found that 5 ammonium carnallite is, depending on the preparing method, extremely little or not at all hygroscopic and resembles e.g. potassium chloride in this respect. The taste of ammonium carnallite is close to the taste of alkali metal chlorides, particularly when the pH of its aqueous solution is arranged close to the pH value of alkali metal chlorides by means of 10 the preparing technique related to the invention. Thus, magnesium double salts related to the invention can be easily added to food salt formulations in such contents that the above-mentioned RDA value can be achieved. It should also be mentioned that thanks to its slightly acid taste, ammonium carnallite prepared under normal conditions is suit-15 able as such or in a mixture with alkali metal chlorides for preparing certain foods, such as salted fish, which normally contains a dangerous quantity of sodium.

As already mentioned above, chlorides, hygroscopic as such, can be easily included in ammonium carnallite to obtain products containing valuable mineral components. Studies indicate that when ammonium carnallite is precipitated from a solution which contains calcium chloride in adjusted ratios, such as in mole ratios of 1/0.1—0.2 of MgCl₂/CaCl₂, a non-hygroscopic salt component is obtained which is pleasant, supplements the taste of the salt product, and which can be used advantageously to make such physiological salt products in which it is desired to include calcium in the formulation.

Furthermore, it has been found that ammonia can be partly replaced by potassium. Thus, a food salt component found to have particular value is a mixed form of ammonium carnallite and potassium carnallite which, depending on the method of preparation, is close to the composition 2MgCl₂ × KCl × NH₄Cl × 6H₂O and which can be preferably manufactured by adding potassium carnallite or, in corresponding ratios, magnesium chloride and potassium chloride, to a mother liquor containing ammonium chloride and magnesium chloride. The ratio of potassium

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and ammonium can be freely selected so that the desired hygroscopic properties are achieved for the product. The hygroscopicity will increase with an increase in the relative proportion of potassium. The carnallite mixtures in question contain exceptionally little ammonium chloride, for whose content in foodstuffs, primarily sweets, an upper limit has been imposed in some countries.

In the general formula of ammonium carnallite, Mg can be partly replaced with calcium, wherein the hygroscopicity will increase with an increase in the proportion of calcium. The general formula of the salt product is thus of the form aMgCl₂ × bCaCl₂ × NH₄Cl × XH₂O, in which the sum of the coefficients a and b is close to the value 1. Consequently, for example a salt product which is made with the a and b values 0.5 and whose composition is 0.5 MgCl₂ × 0.5 CaCl₂ × NH₄Cl × XH₂O, and in which the effect of ammonium carnallite to protect from humidity is weaker, forms, depending on the relative humidity of air, a more or less moist crystalline mass which can, for example in view of the needs of food industry, be dosed in solid form and stored in open vessels without the product being fully deliquesced in the same way as calcium chloride. In this salt form, the content of ammonia is lower, due to the atomic weight of calcium, and the slight hygroscopic nature of the product is not harmful, taking into account the possible use of the product for water binding and seasoning purposes, normally in solution form in cheese and sausage industry and in processed meat industry. In this context, in view of food industry, it should be noted that the ammonium ion contained in the food salt components in question is, due to its slight acidity, detrimental to microbial action. Thus, if these salt products are used instead of sodium chloride, it is possible to reduce correspondingly the quantity of preservatives, for example detrimental nitrite used in sausage industry, and thereby to improve the quality of foodstuffs.

A salt form analogous to the structure of magnesium carnallite, calcium ammonium chloride, $CaNH_4Cl_3 \times 6H_2O$, which corresponds to the particular case a=0 and b=1 of the above-mentioned general structural formula, has excellent taste when compared with calcium chloride $CaCl_2$. The salt is stable when the humidity of air is low, but it has some

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 hygroscopic nature when the humidity of air is high. It has been found that the hygroscopic nature of this salt can be eliminated by modifying it to an organic neutral complex, wherein suitable complexing agents, depending on the desired taste, include hydroxy acids and/or their salts or amino acids and/or their derivatives, preferably glycine. It has also been found that no such disadvantageous changes referring to the oxidation of glycine take place in the taste and colour of the final product, whose structural formula is normally $Gly \times CaNH_4Cl_3 \times H_2O$, which are present in ammonium-free alkaline earth metal complexes of glycine, with reference to the prior art described in Finnish patent applications No. 961229 and No. 970323 (WO 98/32343).

In tests related to the invention, it has been surprisingly found that the properties, such as acidity and thereby taste, of alkaline earth ammonium chlorides, particularly ammonium carnallite, are substantially dependent on the conditions of precipitation, particularly the concentration and pH of the mother liquor. The reason found for this phenomenon is the property of ammonium chloride to be prematurely crystallized from the mother liquor in the presence of slightly acidic salts, such as magnesium chloride, in the solution, which takes place to an increasing extent with an increase in the quantity of water used for the preparation of the mother liquor. Since even relatively small quantities of free ammonium chloride increase the acidity of the double salts, changing their taste to be slightly pungent, the above-mentioned salt product is thus preferably precipitated from a very concentrated solution, possibly under overpressure, wherein the content of free ammonium chloride remains low. The phenomenon is illustrated by means of an example in Fig. 1.

Furthermore, tests related to the invention have surprisingly shown that the acidity, and thereby the taste, of double salt components according to the invention can be substantially influenced by adding small quantities of a base, such as metal hydroxides, preferably ammonium hydroxide or potassium hydroxide, to the mother liquor. The acidity of the liquid can thus be reduced and its pH value be brought to a level at which premature crystallization of ammonium chloride from the mother liquor is prevented. Ammonium hydroxide is a particularly advantageous

alkaline component, thanks to its volatility, but also because no such ions which would complicate the possible re-use of the filtrate will thus be added into the mother liquor. The phenomenon is illustrated graphically by means of an example in Fig. 2.

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Furthermore, tests relating to the invention have also surprisingly shown that for example MgNH₄Cl₃ \times 6H₂O or potassium and/or calcium containing mixed crystal forms of the compound which, treated within the temperature range from 90 to 140°C, lose a part, normally two of their six molecules of water of crystallization, are hydrated by the effect of air humidity to the original stable hexahydrate form whose pH, measured from the aqueous solution, is, depending on the temperature and the time of treatment, higher than the value of untreated material measured in a corresponding way. However, the pH change produced by the thermal treatment normally depends on the hermetic pH value of the product and thus on the method of preparing the product. The phenomenon is illustrated graphically by means of an example in Fig. 3.

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It has been surprisingly found that ammonium carnallite, or mixed crystal forms containing the above compound, produce small quantities of hydrochloric acid HCl with crystal water during thermal treatment and, in a corresponding manner, ammonia NH₃ during a hydration sequence. Thus, the net effect is the discharge of possible coprecipitated ammonium chloride residues from the material, which has also been shown analytically and which also matches with minor weight losses caused by the thermal treatment.

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In this context, it can be mentioned that for example the compound MgNH $_4$ Cl $_3 \times 6$ H $_2$ O is active in a microwave field only if it contains residual moisture. The method can thus be used systematically for drying the products in question without causing changes in the composition of the product.

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As the pH of the ammonium chloride used as a 2 % solution in the tests was 4.53, it is obvious that possible residues of ammonium chloride have an effect on *e.g.* the taste of ammonium carnallite. Therefore, the above-described two methods for controlling the acidity of alkaline earth

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ammonium chlorides in connection with the preparation process have the primary aim to regulate the content of possible free ammonium chloride in the final product and, on the other hand, to have an advantageous effect on *e.g.* the crystal size, filtrability, drying properties, and taste of the product. The optimal pH is, however, determined according to the use of the product and the final salt formulation.

The hydrates which are formed as a result of the thermal treatment and in which the number of molecules of water of crystallization is less than six can be used as such for preparing food salts according to the invention. The hydrate form in question may be advantageous in view of transportation of large quantities of the salt product particularly under damp and warm conditions.

15 Furthermore, it should be stated that such ammonium chloride hydrates of alkaline earth metals, in which the number of molecules of water of crystallization is in the range from 0 to 3, are naturally encompassed by this invention. Their treatment is difficult because of the strong hygroscopic nature of the hydrate forms, and their preparation requires processing at relatively high temperatures, which causes energy consumption and thus additional costs.

In the preparation of the product of the invention, alkaline earth metal chlorides and ammonium chloride, and possibly potassium chloride or calcium chloride and a possible complexing component and possibly a desired quantity of a base, preferably ammonium hydroxide, are brought together as solutions, for example dissolved in water, possibly in equivalent mole ratios, preferably in a concentrated solution at an increased temperature, possibly under overpressure, after which the mixture is mixed to supplement the solubility and cooled at a desired rate to achieve a suitable crystal size. The separated crystal mass is filtered, dried and possibly processed at a temperature range from 90 to 140°C to achieve a hydrate form having approximately four molecules of water of crystallization, is possibly hydrated and is used, as such or mixed with sodium chloride and/or potassium chloride, to prepare a food salt product having the desired ion composition.

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According to a preferred method for preparing the product of the invention, a solution is prepared of magnesium chloride and/or calcium chloride in desired molar ratios, whereby, by mixing with ammonium chloride, a mixed crystal form precipitate is obtained which contains both mineral components as desired. To eliminate the possible hygroscopic nature of the product, the crystal mass is submitted to thermal treatment as described above.

According to a preferred method for preparing the product of the invention, to prepare the alkaline earth component or components for the product, a natural mineral which contains alkaline earth chloride or waste solutions from its processing, for example carnallite $MgCl_2 \times KCl \times 6H_2O$ or waste solutions from its processing, are used.

According to a preferred method for preparing the product of the invention, the affinity of ammonium chloride particularly to magnesium chloride is used, wherein the raw materials to be used can be *e.g.* magnesium chloride waste solutions from potassium chloride industry, normally originating from carnallite and possibly containing alkali bromides; natural deposits, such as carnallite deposits of the Dead Sea; or corresponding solutions which contain the desired mineral components in ratios corresponding to the desired product composition or which can, by suitable processing, made to correspond to the same.

In industrial methods for preparing the product of the invention, it is advantageous to use a continuous process in which *e.g.* an alkaline earth metal ammonium chloride hydrate, or its form containing potassium in addition to ammonium, is brought, after being separated *e.g.* by centrifugation, to a drying line, and the mother liquor is returned to the stage of the process in which it is supplemented *e.g.* with quantities corresponding to the material losses of the raw materials, possibly in the form of solutions, preferably preheated, after which the mixture is concentrated possibly in underpressure, cooled down and brought to the separation stage again. The salt products in question do not form any boiler scale like crusts on the walls of the concentration apparatus, and they are thus well suited for the described process.

One method for preparing the food salt product according to the invention is solid state processing, in which an alkaline earth metal chloride or chlorides and ammonium chloride are brought together in a solid state, possibly together with sodium chloride and/or potassium chloride. To increase the contact surface of the crystals, the mixture is agitated, ground or pulverized *e.g.* in a bead mill, ball mill, or the like, possibly in a continuous process. If necessary, the fine-ground product is allowed to hydrate under normal conditions, after which the product is *e.g.* granulated, when desired.

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The nutritional salt product according to the invention is intended to reduce the daily intake of sodium, whose quantity is considerably high, particularly when industrially prepared and packed foodstuffs are used in Western countries. On the other hand, the food salt product is intended to eliminate possible deficiencies of potassium and magnesium, of which minerals particularly the former has an unquestionable effect reducing the blood pressure. Both of the minerals are primarily intracellular, wherein their need is the greatest during pregnancy, adolescence, sports training, or another stress.

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The components of the physiological salt product of the invention, correcting salting defects in food, can be proportioned within certain limits, for example according to the embodiment examples to be presented hereinbelow, taking into account the new upper limit for the content of potassium chloride set by the Scientific Committee for Food of the EC Commission, which is less than 30 wt-% of the total salt content of the product and which takes into account possible potassium intolerance caused by renal failure.

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To evaluate the medical properties of a salt formulation according to the invention, a test of 5 days was made on five healthy young persons. In the food consumed by the test persons, salt with the following formula was systematically added instead of sodium chloride:

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29% NaCl 36% KCl 35% MgNH₄Cl₃×6H₂O

As a result of the test, it was found that the above-described salt product was absorbed intestinally without side effects and caused a decrease in the average urinal sodium content from the level of 114.8 mmol/l (δ 41.0) to the level of 50.4 mmol/l (δ 32,9) and, correspondingly, an increase in the magnesium content from the level of 4.50 mmol/l (δ 1.78) to the level of 5.48 mmol (δ 2.82). Furthermore, it was significant in the results that the average systolic blood pressure of the test persons decreased from the level of 112.2 mmHg (δ 10.8) to the level of 105.6 mmHg (δ 6.99). In the above values, δ indicates standard deviation.

The following examples describe the technique relating to the invention and some typical salt formulations according to the invention.

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In the examples on preparing alkaline earth metal components, for characterizing the products, the following analytical methods are used:

M g	complexometric, EDTA
NH ₄	Kjeldahl method
CI	AgNO ₃ , adsorption indicator method

The pH of the products is determined from a 2 % aqueous solution.

25 Example 1

This example describes the dependence of the properties of ammonium carnallite as the product on the concentration of the mother liquor.

- a) 1.0 mol (53.5 g) of ammonium chloride was dissolved in 100 ml of water, with heating. The obtained solution was added by mixing to a heated solution which contained 1.0 mol (203.3 g) of the compound $MgCl_2 \times 6H_2O$ dissolved in 50 ml of water. The obtained mixture was cooled, filtered by suction, and dried.
- Yield: 124.4 g (48.5 %) of the compound MgNH₄Cl₃ × 6H₂O Upon evaporation of the mother liquor obtained as a filtrate to half of its original volume, 63.3 g of the same compound was obtained.

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Total yield: 188.2 g /73.3 %), pH 5.27

Analytical values for the product: Mg 8.57 %

NH₄ 8.96 %

Cl 43.8 %

The product contains some coprecipitated ammonium chloride.

b) 1.0 mol (53.5 g) of ammonium chloride and 1.0 mol (203.3 g) of the compound MgCl₂ × 6H₂O were added to 100 ml of water. The mixture was boiled refluxed to complete the solubility and allowed to cool down. The separated crystal mass was filtered by suction and dried.

Yield: 151.2 g (59.0 %), white crystal mass with a pleasant, slightly acid taste.

pH of the product 5.34

Analytical values for the product: NH_4 7.23 %

Mg 9.12 %

Taking into account the error limits of the analysis methods, the product has a composition corresponding to the structural formula MgNH $_4$ Cl $_3 \times 6$ H $_2$ O.

Example 2

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1.0 mol (53.5 g) of ammonium chloride was dissolved by heating to 100 ml of water. When the temperature was 60°C, 1.0 mol (219.0 g) of the compound $CaCl_2 \times 6H_2O$ was added to the mixture. The mixture was cooled, filtered by suction, and dried.

Yield: 110.5 g (40.8 %) of hydrate forms of calcium ammonium chloride When the mother liquor was evaporated to the half of its original volume, 56.5 g of the same product was obtained.

Total yield: 167.0 g (61.7 %)

Analysis: Ca 15.3 %

Example 3

The aim of the example is to describe such a unit process method for preparing ammonium carnallite, in which it is desired to give the product a higher pH value than the product of example 1b).

However, the process of example 1b) was repeated in that 2.5 ml of 5% ammonium hydroxide was added to the mother liquor. The easily filtratable crystal mass was separated from the mother liquor by suction. and the obtained product was dried at a temperature of 60°C.

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Yield: 151.3 g (58.9 %) of cubical crystals and their agglomerates with a pleasant salty taste.

The pH of the product was 5.83 and the analytical values were:

 NH_4

7.50 %

Mg

9.26%

Example 4

The aim of the example is to describe a process of preparing ammonium carnallite in an advantageous manner in industrial scale, in which it is desired to give the product a higher pH than the value of example 3.

107.0 g (2.0 mol) of ammonium chloride and 406.6 g (2.0 mol) of the compound MgCl₂ × 6H₂O were dissolved by heating to 450 ml of water which contained 10 ml of 5 % ammonium hydroxide. The solution was concentrated in underpressure by means of a rotary evaporator until the quantity of distilled water was 290 ml. The separated crystal mass was filtered by suction, dried at room temperature, and the filtrate was 30 recovered to be re-used.

Yield: 345.5 g (61 %) of a crystal mass with a pleasant salty taste. The pH of the product was 6.18 and the analytical values were:

NH₄

7.32 %

Mg

9.23 %

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Example 5

The example describes a situation in which a hygroscopic component is coprecipitated to the ammonium carnallite to be prepared. The product obtained is a calcium-containing non-hygroscopic sprinklable salt component.

 $53.5 \mathrm{~g}$ (1.0 mol) of ammonium chloride, $203.3 \mathrm{~g}$ (1.0 mol) of the compound MgCl₂ \times 6H₂O and 21.9 g (0.1 mol) of the compound CaCl₂ \times 6H₂O were dissolved by heating to 225 ml of water which contained 5 ml of 5 % ammonium hydroxide. The solution was concentrated in underpressure by means of a rotary evaporator until the quantity of the distillate was 145 ml. The separated crystal mass was filtered under suction and dried at a temperature of 70°C.

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Yield: 184.2 g (66 % of the starting materials) of a white crystal mass with a wide salty taste. The pH of the product was 6.44, and the ana-

lytical values were:

NH₄ 6.84 %

Cl 41.60 % Mg 8.52 %

Ca 1.47 %

Example 6

The example describes a situation in which a synthetically formed salt corresponding to natural potassium carnallite is combined with ammonium carnallite, and a non-hygroscopic carnallite mixture is obtained with an exceptionally low NH₄ content.

a) 53.5 g (1.0 mol) of ammonium chloride, 74.5 g (1.0 mol) of potassium chloride, and 406.6 g (2.0 mol) of the compound MgCl₂ × 6H₂O were dissolved by heating into 450 ml of water which contained 5 ml of 5 % ammonia solution. The solution was concentrated in underpressure with a rotary evaporator until the quantity of the distillate was 265 ml.
 The separated crystal mass was filtered by suction and dried at a temperature of 70°C.

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Yield: 390.5 g (73 %) of a white crystal mass with a light salty taste. The pH of the product was 6.10, and the analytical values were:

NH_4	3.55 %
Mg	8.53 %
CI	40.4 %

Taking into account the error limits of the analysis methods, the product corresponds relatively well to the structural formula MgCl₂ \times 0.5 NH₄Cl \times 0.5 KCl \times 6H₂O.

of the distillate was 240 ml. The separated crystal mass was filtered by

b) 26.8 g (0.5 mol) of ammonium chloride, 37.3 g (0.5 mol) of potassium chloride and 203.3 g (1.0 mol) of the compound MgCl₂ × 6H₂O were dissolved by heating into 230 ml of water which contained 2.5 ml of 5 % ammonia solution. To the solution was added the filtrate (165 ml) from the embodiment example 6a, after which the solution was concentrated in underpressure by a rotary evaporator until the quantity

Yield: 320.5 g (78 %) of a salt product with a pleasant taste.

The pH of the product was 5.94, and the analytical values were:

suction and dried at a temperature of 65°C.

NH_4	3.45 %
Mg	9.10 %
CI	39.9 %

Taking into account the error limits of the analysis methods, the product corresponds very well to the structural formula MgCl₂ \times 0.5 NH₄Cl \times 0.5 KCl \times 6H₂O.

30 Example 7

This example describes a situation in which an organic complex is formed of alkaline earth ammonium chloride to have a particularly good absorption capacity.

53.5 g (1.0 mol) of ammonium chloride, 75.1 g (1.0 mol) of glycine, and 219.1 g (1.0 mol) of the compound $CaCl_2 \times 6H_2O$ were dissolved by

heating into 200 ml of water. The solution was concentrated to almost dry state in underpressure by means of a rotary evaporator. The moist crystal mass was carefully dried in a microwave oven.

Yield: 257.2 g of a white crystal mass of small particles corresponding to the formula CaNH₄Cl₃ × Gly × H₂O. The product had a slightly sweet, salty taste. Its pH was 5.47, and the analytical values were:

NH_4	6.73 %
Ca	15.35 %
CI	40.13 %

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Example 8

The example describes a typical Na/K/Mg food salt formulation according to the invention. The middle column shows the amount of the material in milliequivalents with an assumed daily salt intake of 10 g, and the right-hand column shows the daily dose of the corresponding ion in milligrams. The new recommendation on the maximum content of potassium chloride, less than 30 wt-%, is taken into account in the formulation.

	wt-%	mmol	mg
NaCl	36	61.6	1420
KCI	29	38.9	1520
$MgNH_4Cl_3 \times 6H_2O$	35	13.6	330

When the mmol and mg values of the above table are compared with the RDA values of magnesium presented in the introduction of the invention, it can be stated that the physiological objectives set for the invention can be achieved. Thus, the content of magnesium salt in the food salt formulation containing a chloride of sodium and/or potassium is advantageously at least 2.5 wt-%, preferably at least 3.0 wt-%, calculated as magnesium.

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Example 9

The example describes a typical calcium-containing Na/K/Mg food salt product in which the ratio of magnesium and calcium is close to the corresponding average value in sea water. In this example, calcium is present in an easily absorbable complex form. The references in the table are the same as in example 8.

	wt-%	mmol	mg
NaCl	30	51.3	1180
KCI	29	38.9	1520
$MgNH_4Cl_3 \times 6H_2O$	35	13.6	330
$CaNH_4Cl_3 \times Gly \times H_2O$	6	2.33	93

10 **Example 10**

The example describes a typical N/K/Mg food salt product in which the NH₄ content is made exceptionally low by using a mixed form of ammonium and potassium carnallites as the alkaline earth metal component, prepared in mole ratios of 1:1. The references in the table are the same as in the previous examples.

	wt-%	mmol	mg
NaCl	38	65.0	1495
KCI	24	32.2	1260
$MgCl_2 \times 0.5 NH_4Cl \times 0.5 KCl \times 6H_2O$	38	14.2	345

In the example formulation, the content of potassium chloride is reduced, because the carnallite mixture contains 7.1 mmol of potassium. Thus, the total content of KCl becomes 39.3 mmol, which corresponds in the formulation to the percentual content of 29.3 % of KCl, which is less than the present recommendation limit of 30 %.

Claims:

- 1. A physiological food salt product containing an alkaline earth metal component, **characterized** in that said product contains one or more hydrate forms of magnesium ammonium chloride and/or calcium ammonium chloride having the general formula $MNH_4Cl_3 \times XH_2O$, in which formula M is Mg or Ca and X is the number of molecules of water of crystallization.
- 10 2. The product according to claim 1, characterized in that X is within the range from 4 to 6.
 - 3. The product according to claim 1 or 2, **characterized** in that the magnesium ammonium chloride and/or the calcium ammonium chloride is in a complex form.
 - 4. The product according to claim 3, **characterized** in that the complexing compound is a hydroxy-carboxylic acid and/or its salt, or an amino acid and/or its derivative.
 - 5. The product according to claim 1 or 2, **characterized** in that the general anhydrous formula of the salt contained in the product is $aMg \times bCa \times NH_4Cl_3$, in which a+b=1, and a and b are greater than 0, and in which part of the ammonium can be replaced with potassium.
 - 6. The product according to claim 1 or 2, **characterized** in that the general anhydrous formula of the salt contained in the product is in the type $MgNH_4Cl_3 \times eCaCl_2$, in which e is preferably not greater than 0.2 and in which part of the ammonium can be replaced with potassium.
 - 7. The product according to claim 1 or 2, **characterized** in that the general anhydrous formula of the salt contained in the product is Mg \times cNH₄ \times dK \times Cl₃, in which c + d = 1, and c and d are greater than 0, preferably so that c \geq 0.5.
 - 8. The product according to any of the preceding claims, characterized in that it contains sodium chloride and/or potassium chloride.

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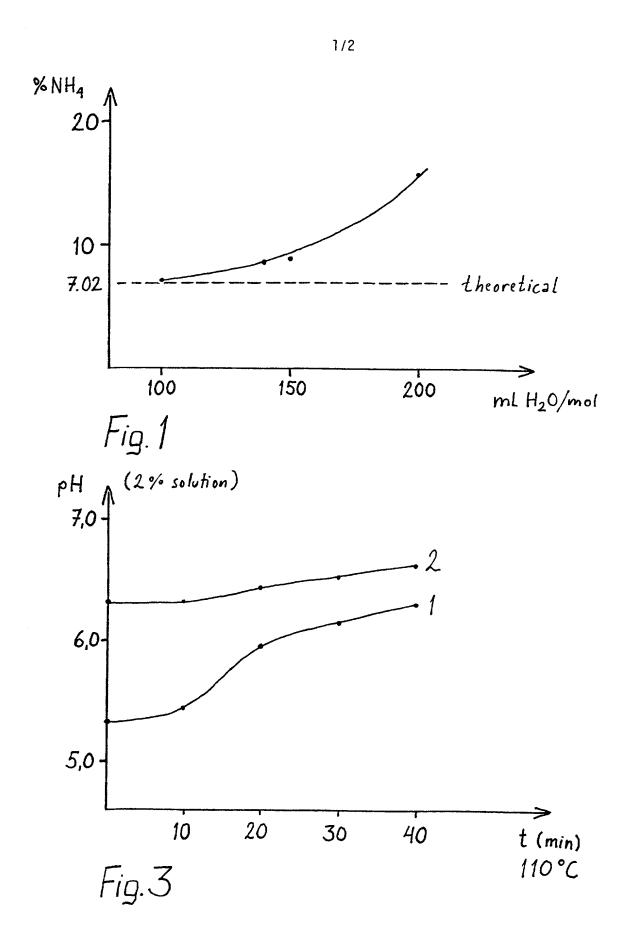
- 9. The product according to claim 8, **characterized** in that the content of magnesium ammonium chloride in the mixture is at least 2.5 wt-%, preferably at least 3.0 wt-%, calculated as magnesium.
- 10. The product according to any of the preceding claims, **characterized** in that it contains materials which are advantageous to vital functions, such as micronutrients, vitamins, flavonoids, steroids, or the like.
- 11. The product according to any of the preceding claims, **characterized** in that it contains as additives affecting primarily the taste of the product carbohydrates or their polymeric forms, spices, herbs, acidity regulators, glutamates, proteins, protein hydrolysates, or the like.
- 12. A nutrient substance, a semi-finished product, a processed food product, a food portion, or the like, **characterized** in that a food salt product according to any of the preceding claims has been used, in solid form or in a solution, in its processing and/or preservation.
- 13. A method for preparing a food salt product containing an alkaline earth metal component, characterized in that an alkaline earth metal chloride and ammonium chloride are brought together in a solution form, wherein a precipitate is formed which contains one or several hydrate forms of an alkaline earth metal ammonium chloride, having the general formula of MNH₄Cl₃ × XH₂O, in which formula M is Mg or Ca and X is the number of molecules of water of crystallization, and the obtained precipitate is separated from the mother liquor.
- 14. The method according to claim 13, **characterized** in that the precipitation is performed in a continuous process, returning the mother liquor after the separation of the precipitate to the stage in which it is supplemented with the alkaline earth metal chloride and ammonium chloride.
- 35 15. The method according to claim 13, characterized in that the solution form contains both magnesium chloride and calcium chloride to include calcium in the product.

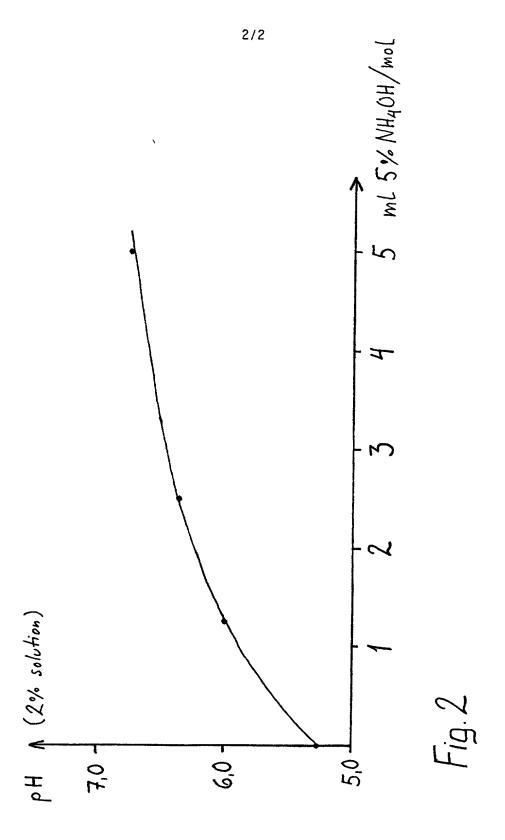
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- 16. The method according to claim 13, **characterized** in that the solution form contains a chloride of potassium, such as KCI, or potassium carnallite MgKCI₃ which also constitutes the alkaline earth metal chloride component.
- 17. The method according to any of the preceding claims 13 to 16, characterized in that the pH of the mother liquor is adjusted by means of a hydroxide, particularly potassium or ammonium hydroxide, particularly to adjust the crystallization of free ammonium chloride.
- 18. A method for preparing a physiological food salt product containing an alkaline earth metal component, **characterized** in that an alkaline earth metal chloride and ammonium chloride are brought together in a solid state possibly together with sodium chloride and/or potassium chloride, and the mixture is agitated, ground, or pulverized *e.g.* in a ball mill or the like, and the obtained product is possibly granulated.





COMBINED DECLARATION AND POWER OF ATTORNEY FOR ORIGINAL, DESIGN, NATIONAL STAGE OF PCT, SUPPLEMENTAL, DIVISIONAL, CONTINUATION OR CONTINUATION-IN-PART APPLICATION

As a below named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated below next to my name,

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled:

the specification of which
a. is attached hereto
b. was filed on as application Serial No and was amended on (if applicable).
PCT FILED APPLICATION ENTERING NATIONAL STAGE
c. was described and claimed in International Application No. <u>PCT/FI00/00018</u> filed on <u>12 January</u> 2000 and as amended on (if any).
I hereby state that I have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment referred to above.
I acknowledge the duty to disclose information which is material to patentability as defined in 37 C.F.R. § 1.56.
I hereby specify the following as the correspondence address to which all communications about this application are to be directed:
SEND CORRESPONDENCE TO: Edward A. Pennington SWIDLER BERLIN SHEREFF FRIEDMAN, LLP 3000 K Street, N.W., Suite 300 Washington, D.C. 20007-5166
DIRECT TELEPHONE CALLS TO: Eric J. Franklin, 202-424-7500
I hereby claim foreign priority benefits under Title 35, United States Code § 119 (a)-(d) or under § 365(b) of any foreign application(s) for patent or inventor's certificate or under § 365(a) of any PCT international application(s) designating at least one country other than the U.S. listed below and also have identified below such foreign application(s) for patent or inventor's certificate or such PCT international application(s) filed by me on the same subject matter having a filing date within twelve (12) months before that of the application on which priority is claimed:

	The attached 35 U. declaration.	S.C. § 119 claim for p	priority for the applicati	ion(s) listed below for	orms a part of this
	Country/PCT	Application Number	Date of filing (day, month, yr)	Date of issue (day, month, yr)	Priority Claimed
	Finland	990145	27 January 1999		XY N NY N
					YN
	I hereby claim the l	penefit under 35 U.S.	C. § 119(e) of any U.S.	provisional applicat	ion(s) listed below.
	Provisional Appl	ication No.	Date of fi	ling (day, month, yr)	
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A			IONAL, CONTINUAT APPLICATION(S DE		
			States Code § 120 of an signating the U.S. lister		lication(s) or under §
US/PO	CT Application Serial	No. Filin		Etatus (patented, pen U.S. application no.	
US/PO	CT Application Serial	No. Filir		Status (patented, pen U.S. application no. a	
In this continuation-in-part application, insofar as the subject matter of any of the claims of this application is not disclosed in the above listed prior United States or PCT international application(s) in the manner provided by the first paragraph of Title 35, United States Code, § 112, I acknowledge the duty to disclose material information as defined in Title 37, Code of Federal Regulations, § 1.56(a) which occurred between the filing date of the prior application(s) and the national or PCT international filing date of this application.					
inform willfu Title	nation and belief are b I false statements and	elieved to be true; an the like so made are a Code and that such		ements were made wnprisonment, or both	vith the knowledge that a, under Section 1001 of



I hereby appoint the following attorneys and/or agents with full power of substitution and revocation, to prosecute this application, to receive the patent, and to transact all business in the Patent and Trademark Office connected therewith: Edward A. Pennington (Reg. No. 32,588), John P. Moran (Reg. No. 30,906), Eric J. Franklin (Reg. No. 37,134), Michael A. Schwartz (Reg. No. 40,161), Robert C. Bertin (Reg. No. 41,488), Alicia A. Meros (Reg. No. 44,937), Chadwick A. Jackson (Reg., No. 46,495), Edward J. Naidich (Reg. No. 43,826), and Sean O'Hanlon (Reg. No. 47,252) of Swidler Berlin Shereff Friedman having an address of 3000 K Street, N.W., Suite 300, Washington, D.C. 20007-5116.

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Full name of sole or first inventor Juhani MÄKI Inventor's signature*

Residence: Kaivanto, Finland

Citizenship: Finland

XIZ

Post Office Address: Vanha Pälkäneentie 157, FIN-36570 Kaivanto, Finland

Full name of second inventor

Inventor's signature*

date

date 13 July 2001

Residence: Citizenship:

Post Office Address:

Full name of third inventor

Inventor's signature*

date

Residence: Citizenship:

Post Office Address:

Full name of fourth inventor

Inventor's signature*

date

Residence: Citizenship:

Post Office Address: